

ESR Spectrometry of High-Temperature Superconductors with Temperature-Modulation-Based Thermoregulation

M.K. Aliev, G.R. Alimov¹, I.Kholbaev,
T.M. Muminov, B. Olimov, B.Yu. Sokolov,
and R.R. Usmanov,

Institute of Applied Physics, Tashkent State University 700095, Tashkent, Uzbekistan

Abstract

A light-beam-assisted temperature-control system operating within a temperature range from 77 to 180 K is developed for investigating the absorption of electromagnetic waves by high-temperature superconductors in the vicinity of superconducting transition with an ESR spectrometer. The advantage of this system is the feasibility of modulating the temperature of a sample with a frequency of 80 Hz and amplitude of $10^{-2} \div 10^{-1} K$. The rms temperature instability over a 5-min time interval is within 0.06 K, the temperature gradient in a sample is $\sim 10^{-2} K/mm$ for $T \sim 90 K$, and the system relaxation time is 1 – 10 s. The microwave absorption peaks in the vicinity of the superconducting transition of an $YBa_2Cu_3O_{7-x}$, single crystal were first measured at (the simultaneous application of magnetic and temperature modulations).

Keywords: Temperature modulation; Magnetic-field modulation; Superconducting transition; Microwave absorption; High- T_c superconductor

1 Introduction

It is known that the temperature dependence of microwave response in high-temperature superconductors (HTSCs), which is recorded by ESR spectrometers using the magnetic modulation technique, peaks at the superconducting transition temperature [1] - [4]. This signal can serve as a source of information on the features of the superconducting transition in HTSCs. The observation of this signal, when temperature modulation is used, and its comparison with the signal recorded with the use of magnetic modulation are of significant interest. Note that ESR spectrometers with flow-type cryostats [5] do not provide the necessary temperature uniformity over a sample [6] and, in addition, it is difficult to modulate the temperature of a sample in them.

Below we describe a temperature-control system designed to study HTSCs at an SE/X 2543 RADIOPAN ESR spectrometer. The sample temperature is controlled with a light beam. This system has significantly better

¹Corresponding author. e-mail: gleb@iaph.silk.org

characteristics in a temperature range of $77 < T < 120K$ than flow-type cryostats, and, importantly, allows us to modulate the sample temperature with a required frequency.

2 Method

The left part of Fig. 1 shows the general view of our cryosystem. Figure I (central part) shows the design of the sample holder (SH), which is one of the most important elements that determine technical parameters of the cryosystem.

The working volume of the holder is represented by a thin-walled capsule 1. It is placed inside a fingerlike extension of a quartz dewar (D) with liquid nitrogen, which is inserted into the microwave resonator (MR) through an opening in its top wall. The SH dimensions are determined mainly by those of the resonator and dewar D.

Sample 2 and the working junction of a differential chromel-copel thermocouple 3 (wires have a diameter of 0.15 mm) being inside the capsule are placed inside heat exchanger 4. The latter is a sapphire cylinder sawed into two parts, as shown in Fig. 1. The main, larger part of the heat exchanger has depressions, into which the working junction of the thermocouple with wires is pressed. To ensure a better thermal contact with the heat exchanger, hollow cavities are filled with a mixture of a finely dispersed corundum powder with a silicate adhesive.

The reference junction of the thermocouple is placed in a separate dewar with liquid nitrogen (not shown in Fig. 1), where its temperature is monitored to an accuracy of $\pm 0.005K$ with a 650H UNIPAN platinum resistance thermometer. The chromel-copel thermocouple has a sensitivity of $\geq 30\mu/K$ at the liquid nitrogen temperatures. This makes it possible to monitor the temperature variation for the working junction of the thermocouple to an accuracy of $\pm 0.03K$.

The main part of the heat exchanger has a saw cut 0.3 mm wide and 1 mm deep along the cylinder diameter, in which the sample under study is mounted (as a rule, samples of HTSC single crystals have the form of plates with a width of a few tens of microns). The sample is separated from the working junction of the thermocouple in order to avoid the influence of local distortions of the microwave field caused by the junction. A high thermal conduction of sapphire at low temperatures provides the correspondence between the thermocouple readings and sample temperature to a fairly high

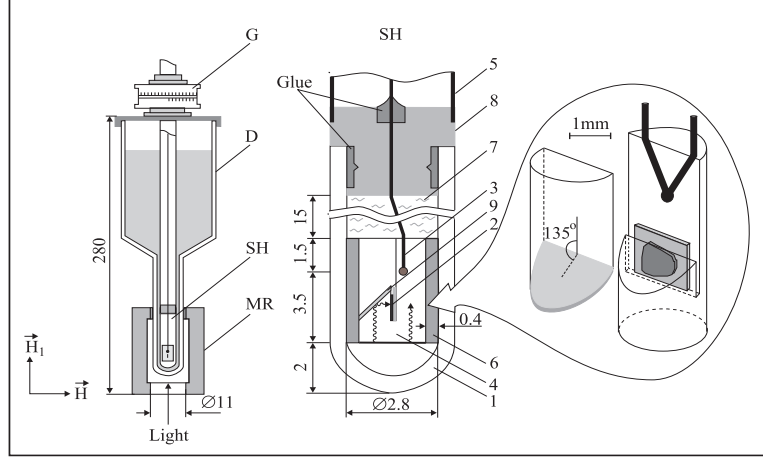


Fig.1. Designs of the cryosystem and sample holder: (1) quartz capsule; (2) sample; (3) thermocouple junction; (4) sapphire heat exchanger; (5) rod of the sample holder; (6) heat insulator; (7) cotton layer; (8) ebonite plug; (9) reflector of the heat wave; (D) dewar; (SH) sample holder; (MR) microwave resonator; and (G) goniometer.

accuracy (the temperature difference between the sample and thermocouple junction evaluated below is within 0.05 K for T-90 K). A sample is glued to a backing of a dense paper with the thickness that provides mechanical pressing of the sample plane against the heat exchanger (against the left wall of the saw cut in Fig. 1). To create a reliable thermal contact with the heat exchanger, a thin silicone grease layer is deposited on the sample surface.

The sample plane is oriented along the direction of the magnetic component \mathbf{H}_1 , of the microwave field. Such an orientation of the sample is optimum for the study of anisotropic effects, because this allows us to vary the angle between the sample plane and external magnetic field \mathbf{H} within the maximal possible range by rotating the capsule about its axis. Rod 5 of the sample holder (a thin-walled tube made of stainless steel) is fixed by a collet type clip of the goniometric head (G), thus making it possible to turn the capsule around the vertical axis through an arbitrary angle.

The sample is heated to a temperature above the nitrogen one by the light flux, which is focused on the blackened bottom end of the heat exchanger by using a system of lenses. A KGM-150 halogen lamp serves as a light source. Its power and, consequently, the light flux intensity incident on the heat exchanger is controlled with the help of a regulated dc power supply. The minimal step in temperature variations is within 0.06 K.

The efficiency of sample heating depends on the heat insulator 6 material used, which plays the principal role in the heat exchange between the sapphire

cylinder and thermostat (liquid nitrogen N_2). The quality of thermal contact can be varied by changing the heat insulator material. A layer of fluffed up cotton 7 filling the space between the heat exchanger and ebonite plug 8 of capsule 1 prevents from the appearance of thermal convective air flows inside the capsule. When cotton was used as a heat insulator 6, the sample temperature reached a value of 180 K at the maximum lamp power.

The stability of the sample temperature at a given current through the lamp filament is strongly dependent on the capsule sealing and liquid nitrogen purity. As was experimentally noted, the instability caused by liquid nitrogen overheating is efficiently removed by a well-known method of using a cotton thread immersed into liquid nitrogen down to the level of the bottom end of capsule 1. If the appropriate conditions were satisfied, the rms instability of the sample temperature was within 0.06 K over a 5-min interval.

The sample temperature modulation is implemented by 80-Hz chopping of the light flux with a two-lobe obturator, which also controls the operation of an optical sensor (photodiode-light-emitting diode) generating the reference signal for the synchrodetector of the receiving and measuring circuit. The choice of this frequency is connected with an appropriately modernized 80 Hz UNIT block, which is included in the ESR spectrometer set and normally used for low-frequency magnetic modulation with a frequency of 80 Hz. Here, this block serves as a synchrodetector.

An IBM PC/AT computer connected to the CAMAC bus controls the system operation, monitors its parameters, and performs the acquisition, processing, and visualization of the data obtained.

Owing to the simplicity of the thermoregulating system, its most important characteristics can be determined by computations. Below we give the appropriate estimates obtained by using the reference data from [7] - [8].

To evaluate the heat flux $W_{in}(T_{av})$ conveyed to the bottom end of the sapphire cylinder for maintaining its average temperature T_{av} , we assume that because of a high thermal conductivity of sapphire ($\lambda \approx 700 W/m \cdot K$) for $T \approx 90 K$, the temperature at an arbitrary point of the cylinder is close to its average value. Then, in view of the fact that under the steady-state conditions $W_{in}(T_{av})$ is equal to the heat flux $W_{out}(T_{av})$ leaving the cylinder through its surface, we can write

$$W_{in}(T_{av}) = W_{out}(T_{av}) \approx \frac{\lambda' S}{\delta} (T_{av} - T_a), \quad (1)$$

where λ' is the thermal conductivity of the heat insulator in the gap between the lateral surface of the cylinder and the capsule wall, δ is the gap value, S

is the area of the lateral surface of the cylinder, and T_a is the temperature of the internal surface of the capsule that, according to crude estimates, can be assumed equal to the temperature of liquid nitrogen. On the right-hand side of the latter equality, we omitted the terms that correspond to the heat fluxes leaving the cylinder through its upper end and through the thermocouple wires, because their overestimated total contribution is no larger than 10%. The factor $\alpha \equiv \lambda' S / \delta$ for the heat insulator in the form of paper ($\lambda' \approx 4 \cdot 10^{-2} \text{W/m} \cdot \text{K}$) and air $\alpha \approx 3 \cdot 10^{-3} \text{W/K}$ takes on the value ($\lambda' \approx 10^{-2} \text{W/m} \cdot \text{K}$) and $\alpha \approx 8 \cdot 10^{-4} \text{W/K}$, respectively.

Now it is easy to assess the uniformity of the temperature distribution over the sapphire cylinder volume. Taking into account that the heat flux density and, consequently, the temperature gradient $|\nabla T|$ inside the cylinder volume is maximum near the bottom cylinder end, we can write the inequality

$$|\nabla T| \leq \frac{W_{in}(T_{av})}{\lambda \sigma} \approx \frac{\alpha}{\lambda \sigma} (T_{av} - T_a), \quad (2)$$

where σ is the area of the cylinder end and λ is the thermal conductivity of sapphire. In contrast to the heat insulator, the thermal conductivity of sapphire strongly depends on the temperature in the region we are interested in and changes from $\lambda \sim 10^3 \text{W/m} \cdot \text{K}$ for $T \sim 80 \text{K}$ to $\lambda \sim 10^2 \text{W/m} \cdot \text{K}$ for $T \sim 120 \text{K}$. From ((2)) it follows that for a paper heat insulator, $|\nabla T| \leq 2 \cdot 10^{-2} \text{K/mm}$ for $T_{av} \approx 90 \text{K}$ and $\leq 5 \cdot 10^{-1} \text{K/mm}$ for $T_{av} \approx 120 \text{K}$, and in case the air is used for heat insulation, $|\nabla T| \leq 6 \cdot 10^{-3} \text{K/mm}$ for $T_{av} \approx 90 \text{K}$ and $|\nabla T| \leq 10^{-1} \text{K/mm}$ for $T_{av} \approx 120 \text{K}$. We note for comparison that in the best flow-type cryostats used as temperature attachments to ESR spectrometers, the temperature gradient at the location of a sample can reach a value of 0.5 K/mm within the temperature range examined [6]. The temperature difference in a sample with linear dimensions of $\sim 1 \text{ mm}$ that corresponds to this value turns out to be comparable with the width of the superconducting transition $\Delta T \sim 0,5 - 1 \text{K}$, which is characteristic of HTSC 1-2-3 single crystals with the critical temperature $T_c \sim 90 \text{K}$. It is obvious that the shape of the superconducting transition peak in this situation must be strongly distorted in comparison with the true one.

The time of establishing a steady-state temperature field in the capsule at a fixed current through the lamp filament is governed mainly by the time of transition to a steady state in the sapphire-thermostat system. This time can be expressed in the known form [9]

$$t_r = \frac{cm}{\alpha},$$

where c is the specific heat for sapphire and $m=60$ mg is the cylinder mass. For $T=90$ K ($c \approx 100 J/kg \cdot K$), we have $t_r \approx 2c$ for paper as a heat insulator and $t_r \approx 8c$ for the air as a heat insulator. The measurements performed have shown the correctness of these estimates. Note that in flow-type cryostats, the time of establishing a preset temperature of a sample is a few minutes.

When a light beam is chopped with the frequency ν , the time dependence of the heat flux $W_{in}(t)$ transferred to the cylinder has a meander form, so that the following expansion is valid:

$$W_{in}(t) = W_0 + \frac{4W_0}{\pi}(\cos \omega t - \frac{1}{3} \cos 3\omega t + \dots),$$

where $\omega = 2\pi\nu$. The constant component W_0 of this expansion has the physical meaning identical with that of $W_{in}(T_{av})$ in (1), i.e., $W_0 = W_{in}(T_{av})$. In this case, T_{av} denotes the cylinder temperature averaged not only over the volume, but over time as well. The amplitude of the fraction of the flux that oscillates with its fundamental frequency can be found in the form $W_\omega = 4/\pi W_{in}(T_{av})$. This allows us to evaluate the amplitude of the temperature wave $\tau_0 \exp[i(kx - \omega t)]$ generated in the direction of the cylinder axis by using the known formula [10]

$$\tau_0 = -\frac{W_\omega}{\sigma \lambda \cdot ik} = \frac{4 W_{in}(T_{av}) \cdot i}{\pi \sigma \lambda k},$$

where $k = (1 + i)(c\rho\omega/2\lambda)^{1/2}$ is the complex wavenumber (ρ is the sapphire density). For $T_{av} \sim 90K$, we have $|\tau_0| \approx 4 \cdot 10^{-2}K$ for a paper heat insulator and $|\tau_0| \approx 10^{-2}K$ for the air heat insulator. It should be noted that as a result of damping, at the level of the sample location the wave is expected to have smaller amplitudes than indicated above, because the penetration depth for sapphire is $l = (\text{Im } k)^{-1} \approx 2mm$, which slightly exceeds the distance from the bottom end of the cylinder to the sample. Moreover, it was observed experimentally that the temperature modulation amplitude in thin plates of HTSC single crystals with the width $d < 0,05mm$ is significantly dependent on the angle between the plate plane and the direction of wave propagation in sapphire, i.e., the angle of slide: the height of the peak of the superconducting transition, from which the temperature modulation amplitude was evaluated, was at a noise level for zero angles and rapidly increased with the angle increase. A small modulation amplitude at zero angles of slide means that in these cases the amplitude of the temperature wave entering the sample is suppressed. As a consequence, we infer that the orientation of the sample plane along the cylinder axis used

in experiments is less favorable from the viewpoint of creating a temperature modulation and conserving the initial direction of the temperature wave. This difficulty is eliminated by reflector 9 (Fig. 1), Its role is played by the boundary between sapphire and cardboard spacer, the thermal conductivities of which have a ratio of $\sim 10^3$. The cardboard spacer has a much larger thickness ($d \approx 0,1mm$) than the penetration depth (for cardboard, $l=0.02$ mm) and guarantees that the reflectance is close to unity. The reflector plane makes an angle of 45° with the cylinder axis and provides not only the normal incidence of the reflected wave on the sample surface, but also a high uniformity of the amplitude and phase of the temperature modulation along this plane, because all the imaginary rays reaching the sample after being reflected have identical path lengths (a wavy line with an arrow in Fig. 1 shows the direction of the temperature wave propagation). The temperature modulation uniformity over the sample thickness is achieved by using another reflector, represented by the boundary with a paper spacer that presses a sample against sapphire.

It can be demonstrated that the reflected wave, which originates due to a strong decrease in the heat conduction at the sample-paper interface, interferes with the primary wave and must effectively smooth the amplitude and phase of temperature oscillations inside the sample. This statement is valid only for the samples, the thickness of which is small enough in comparison with the penetration depth (as a rule, HTSC single crystals with the thickness $d < 0.05mm$ satisfy this requirement). Because of a large decrease in the heat conduction at the sapphire-sample interface, the amplitude of the resulting wave inside the sample must exceed that of the wave incident onto this interface by a factor of almost 2. This gain satisfactorily compensates for the temperature wave damping on the way to the sample. Therefore, the above-mentioned values of $|\tau_0|$ can be used as estimates for the temperature modulation amplitude in the sample.

3 Results

To illustrate the functional feasibilities of the cryosystem described, Fig. 2 presents the results of measuring the first derivative of the microwave power R absorbed by an $YBa_2Cu_3O_{7-x}$ single crystal measuring $\sim 1,0 \times 0,7 \times 0,03mm$. The results were obtained according to the conventional technique with the modulation of an external magnetic field strength (the frequency is 100 kHz and modulation amplitude is 10 Oe) and with our

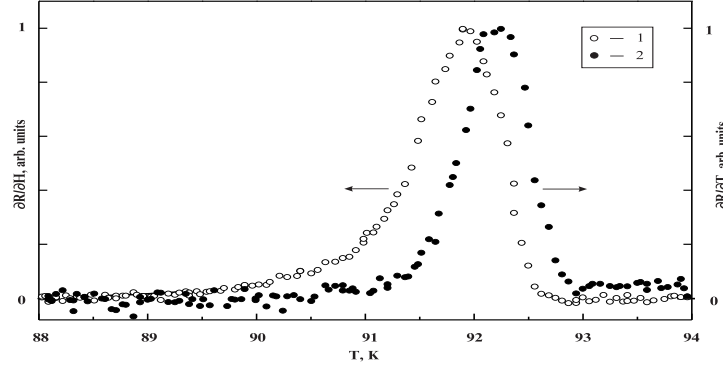


Fig.2. Temperature-dependent microwave absorption in an $YBa_2Cu_3O_{7-x}$, single crystal measured by the (1) magnetic and (2) temperature modulation methods.

method using the sample temperature modulation (the frequency is 80 Hz and modulation amplitude is $10^{-2} \div 10^{-1} K$). Measurements were performed under the conditions of cooling the sample from above-critical temperatures in the external field $H = 20$ Oe directed perpendicular to the sample plane.

In summary it should be noted that the initial, less perfect version of this system of thermal regulation was applied to the investigations of microwave absorption in HTSC ceramics [2].

References

- [1] Moorjani K., Bohandy J., Adrian F.J., et al., Phys. Rev. B: Condensed Matter, 1987, vol. 36, p. 4036; Kim B.F., Moorjani K., Bohandy J., and Adrian F.J., J. Appl. Phys., 1988, vol. 63, p. 2029.
- [2] Aliev M.K., Vavryshchuk Ya., Volosyanyi S.P., et al., Preprint of the Joint Inst. for Nuclear Research, Dubna, 1988, no. R14-88-477; Aliev, M.K., Vavryshchuk, Ya., Volosyanyi, S.P., et al., Fiz. Tverd. Tela (Leningrad), 1989, vol. 31, no. 9, p. 254.
- [3] Shaltiel D., Bill H., Crayevsky A., et al., Phys. Rev. B: Condensed Matter, 1991, vol. 43, no. 16, p. 13594.
- [4] L'vov S.G., Talanov Yu.I., Khasanov R.I., and Shustov V.A., Sverkhprovodimost: Fiz., Khim., Tekn., 1993, vol. 6, no. 6, p. 1175.

- [5] Belyaeva A.I., Silaev V.I., and Stetsenko Yu.V., Protochnye kriostaty dlya laboratornykh issledovaniy (Flow-Type Cryostats for Laboratory Investigations), Kiev: Naukova Dumka, 1987.
- [6] Goloshchapov S.I., Veinger A.I., and Konnikov S.G., Prib. Tekh. Eksp., 1993, no. 3, p. 232.
- [7] Spravochnik po fiziko-tekhnikeskim osnovam kriogeniki (Handbook on Physicotechnical Foundations of Cryogenics), Malkov M.P., Ed., Moscow: Energoatomizdat, 1985.
- [8] Tablitsy fizicheskikh velichin (Tables of Physical Quantities), Kikoin I.K., Ed., Moscow: Atomizdat, 1976, p. 1008.
- [9] Sullivan P.F. and Seidel G., Phys. Rev., 1968, vol. 173, no. 3, p. 679.
- [10] Lykov A.V., Teplomassoobmen (Handbook) [Heat Exchange (Handbook)], Moscow: Energiya, 1978, p. 147.